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ThermoCalc Modelling of the Effect of Chemical Composition on the Phase Transformations Temperatures of Ti-0.4Al Alloy

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Abstract. The effect of chemical composition of Ti-0.4Al α -titanium alloy (PT-1M alloy) on the phase transformation temperatures up to melting temperature has been investigated using ThermoCalc software. The possibility of appearance of Ti_2N nitride, $Ti_x(CN)_y$ -type carbonitride, and Ti_2Fe -type intermetallic in the alloy structure in equilibrium state has been proved by thermodynamic calculations. It has been proposed the so-called aluminum equivalent (Al_{eq}) for PT-1M alloy to analyze the effect of chemical composition on the phase transformation temperatures change. The following equivalents have been suggested: $Al_{eq}^{T\beta}$ for $\beta \leftrightarrow (\beta + \alpha)$ – transformation, $Al_{eq}^{T_{liq}}$ for $L \leftrightarrow \beta$ – transformation, and $Al_{eq}^{T_{sol}}$ for $(L + \beta) \leftrightarrow \beta$ – transformation. It has been shown the linear correlation between the characteristic temperatures of phase transformations in the investigated alloy and the proposed aluminum equivalents.

1. Introduction

Ti-0.4Al alloy (also known as PT-1M alloy) refers to a low-alloyed α -titanium alloys [1]. This alloy is characterized by good technological plasticity both in hot and cold state, high corrosion resistance, and moderate strength [2]. PT-1M alloy is used for manufacture of deformable semi-finished products (sheets, rods, pipes etc.) for products operating in sea and fresh water with operating temperatures up to 150 °C [3, 4]. The change in the chemical composition varying within the alloy grade of PT-1M alloy should affect the phase transformations temperatures: liquidus temperature (T_{liq}), solidus temperature (T_{sol}), $\beta \leftrightarrow \alpha + \beta$ – polymorphic transformation temperature etc. These phase transformation temperatures are important for a reasonable choice of processing parameters of casting, forming, and heat treatment [5, 6, 7]. One of the promising ways to obtain the phase transformations temperatures is a thermodynamic calculation using ThermoCalc software [8, 9]. However, there is a lack of information about the influence of chemical composition of PT-1M alloy on the phase transformation temperatures in this alloy up to melting temperature. The main purpose of the present study was to calculate the phase transformation temperatures of PT-1M titanium alloy with different chemical composition varying within the alloy grade, as well as to estimate the effect of chemical composition on these temperatures.

2. Experimental

The chemical composition of PT-1M alloy is Ti-0.2...0.7 Al-0.2 Fe-0.3 Zr-0.1 Si-0.12 O-0.07 C-0.04 N-0.006 H (wt.%), where Al is an alloying element, and Fe, Zr, Si, O, C, N, and H are impurities according to GOST 19087-91 standard.



Several chemical compositions varying within the alloy grade were chosen for the investigation. Selected compositions provided different combinations of α -stabilizers (Al, O, N, C) and β -stabilizers (Fe, Si) such as: $\min \alpha + \max \beta$, $\min \alpha + \min \beta$, $\max \alpha + \max \beta$, $\max \alpha + \min \beta$, where $\min \alpha$ and $\max \alpha$ is minimum and maximum content of α -stabilizers, relatively; $\min \beta$ and $\max \beta$ is minimum and maximum content of β - stabilizers, correspondingly. Minimum and maximum content of Al as well as maximum content of impurities were taken from GOST standard. Minimum content of impurities was taken as 1/10 of maximum content set up by GOST standard. Zirconium was considered as a neutral element. The possible presence of hydrogen was not taken into account due to its low content.

Two additional chemical compositions of PT-1M alloy (average grade composition and composition used in [9,10]) were also studied. The chemical composition of all investigated alloys is given in Table 1.

Table 1. Chemical composition of investigated alloys (wt.%).

| Alloy # | Al | Zr | Si | Fe | O | N | C |
|--------------------------------------|------|-------|-------|------|-------|-------|--------|
| Alloy 1 – $\min \alpha + \max \beta$ | 0.2 | 0.3 | 0.1 | 0.2 | 0.012 | 0.004 | 0.007 |
| Alloy 2 – $\max \alpha + \min \beta$ | 0.7 | 0.03 | 0.01 | 0.02 | 0.12 | 0.04 | 0.07 |
| Alloy 3 – $\max \alpha + \max \beta$ | 0.7 | 0.3 | 0.1 | 0.2 | 0.12 | 0.04 | 0.07 |
| Alloy 4 – $\min \alpha + \min \beta$ | 0.2 | 0.03 | 0.01 | 0.02 | 0.012 | 0.004 | 0.007 |
| Alloy 5 – Average grade composition | 0.45 | 0.165 | 0.055 | 0.11 | 0.066 | 0.022 | 0.0385 |
| Alloy 6 – Melt composition | 0.4 | – | – | 0.04 | 0.08 | 0.01 | 0.03 |

Temperatures of phase transitions were calculated using ThermoCalc software. This software is based on model of classical thermodynamics, which deals with systems in equilibrium state [11].

3. Results and discussion

The dependence of change in the number of phases in all investigated alloys in the temperature range from 100°C to 2000°C has been obtained using ThermoCalc software. The calculation results for Alloy 4 ($\min \alpha + \min \beta$) and Alloy 3 ($\max \alpha + \max \beta$) are given in Figure 1.

Based on the calculation results it has been determined the thermodynamically probable temperatures of possible phase transitions such as liquidus temperature ($T_{\beta \leftrightarrow L}$), solidus temperature ($T_{\beta \leftrightarrow L + \beta}$), $\beta \leftrightarrow \beta + \alpha$ – polymorphic transformation temperature (T_{β}), and $\beta + \alpha \rightarrow \alpha$ – polymorphic transformation temperature ($T_{\beta + \alpha \rightarrow \alpha}$), as well as onset temperatures of forming of titanium carbide (T_{TiC}), Ti_2Fe -type intermetallic (T_{Ti_2Fe}), and Ti_2N nitride (T_{Ti_2N}). The obtained temperatures for all examined alloys are given in Table 2.

Table 2. Temperatures of phase transitions of investigated alloys.

| Alloy # | $T_{\alpha + Ti_2N \leftrightarrow \alpha},$ °C | $T_{\alpha + Ti_2C \leftrightarrow \alpha},$ °C | $T_{\alpha + Ti_2Fe \leftrightarrow \alpha},$ °C | $T_{\alpha + Ti_2Fe}$ $\leftrightarrow \beta + \alpha,$ °C | $T_{\alpha \leftrightarrow \beta + \alpha},$ °C | $T_{\beta + \alpha \leftrightarrow \beta},$ °C | $T_{\beta \leftrightarrow L + \beta},$ °C | $T_{\beta + L \leftrightarrow L},$ °C |
|---------|--|--|---|--|--|---|--|--|
| Alloy 1 | – | 341 | – | 546 | – | 886 | 1635 | 1677 |
| Alloy 2 | 151 | 646 | 409 | – | 837 | 949 | 1672 | 1745 |
| Alloy 3 | 153 | 663 | – | 553 | – | 946 | 1640 | 1735 |
| Alloy 4 | – | 339 | 375 | – | 825 | 892 | 1665 | 1681 |
| Alloy 5 | – | 530 | – | 546 | – | 920 | 1653 | 1715 |
| Alloy 6 | – | 478 | 505 | – | 724 | 913 | 1671 | 1697 |

As it can be seen from Table 2 and Figure 1, the calculations point out the possibility of appearance in the alloy structure of such equilibrium phases as Ti_2N nitride with HCP lattice, titanium carbide (Ti_2C) with FCC lattice, and Laves phase on the base of Ti_2Fe intermetallic.

According to the calculations, their equilibrium content does not exceed tenths of a percentage point. The analysis of data given in Table 2 showed that the stability of these phases is directly depends on the content of nitrogen, carbon, and iron for Ti_2N , Ti_xC , and Ti_2Fe intermetallic, correspondingly.

There is a lack of information about the formation of above-mentioned phases in PT-1M alloy. It is reported [12, 13] that formation of Ti_2Fe intermetallic occurs in a commercial pure titanium (which is close to investigated PT-1M alloy) upon long tempering (approximately 500 h) in the temperature range from 300 °C to 600 °C. Formation of Ti_2Fe is a result of the appearance on the grain boundaries the micro segregations enriched with iron [12, 13]. The temperature range of formation of Ti_2Fe intermetallic calculated for PT-1M alloy in the present study is from 375 °C to 553 °C (Table 2) and corresponds well to a previously mentioned data.

According to ThermoCalc calculations, Ti_2Fe intermetallic contains mainly titanium and iron, and lesser zirconium. The ratio of these elements gives $(TiZr)_xFe$ compound, where $x=2.3$, i.e. close to stoichiometry of Ti_2Fe compound. It is reported about the possibility of simultaneous enrichment with zirconium and iron of grain boundary areas in Ti-Al-Zr alloy with Fe impurity [14].

It should be noted that in PT-1M alloy containing more than 0.11% Fe (wt.%) the formation of Ti_2Fe intermetallic occurs at substantially constant temperature of 550 ± 4 °C by $\alpha + Ti_2Fe \leftrightarrow \beta$ eutectoid reaction. The calculation results showed that in the course of this reaction the alloy containing more than 0.11% Fe (wt.%) does not turn into single α -phase state during cooling. Therefore, the temperature of $\beta \leftrightarrow (\beta + \alpha)$ – polymorphic transformation ($T_{\beta \rightarrow \alpha}$) cannot be calculated (Table 2).

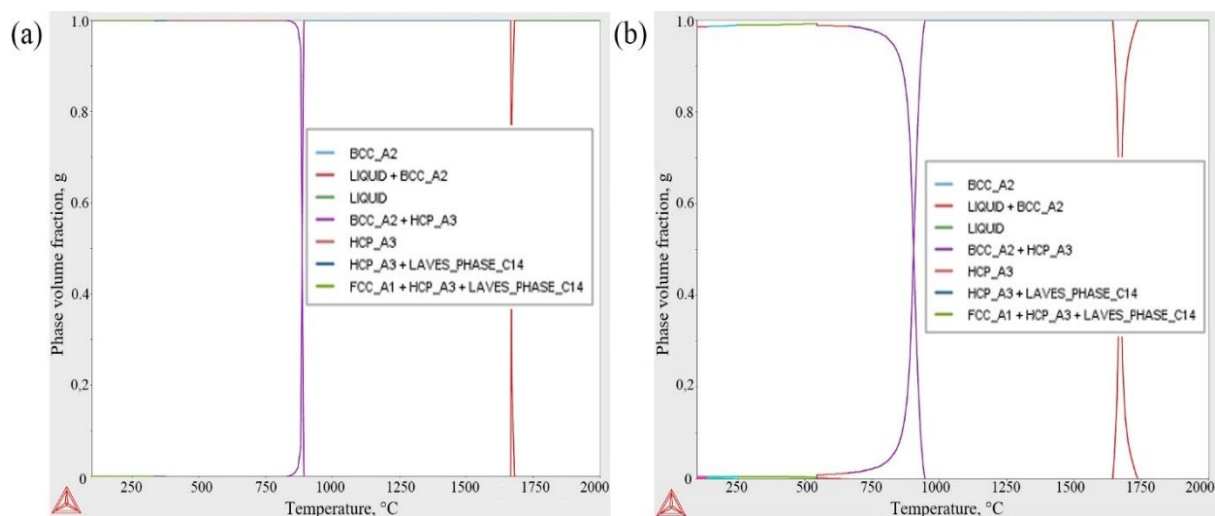


Figure 1. Temperature dependence of volume fraction of phases in Alloy 4 (min α + min β) (a) and Alloy 3 (max α + max β) (b) obtained by use ThermoCalc software, where LIQUID is liquid state, BCC_A2 is titanium β -solid solution with BCC lattice, HCP_A3 is titanium α -solid solution with HCP lattice, LAVES_PHASE_C14 is Laves phase on the base of Ti_2Fe intermetallic, FCC_A1 is phase on the base of titanium carbide with FCC lattice, Ti_2N is titanium nitride.

Lowering of iron content from 0.02 wt.% to 0.04 wt.% in PT-1M alloy results in Ti_2Fe -type intermetallic precipitation from α -phase, and decrease in onset temperature of precipitation from 505°C to 375°C. This is consistent with published data [15] where it is shown that in Ti-Fe alloy the precipitation of intermetallic phase is possible at iron content higher than 0.01 wt.%.

It was reported in [16] that carbide phase was observed in a commercial pure titanium. It was noted that usually this phase is not a carbide, but a carbonitride. The calculated chemical composition of the carbide in PT-1M alloy [9,10] confirms the above data. According to ThermoCalc calculations, the obtained equilibrium phase is $Ti_x(CN)$ carbonitride, where $x = 1.64$, and C/N ratio equals 10/1. The

stoichiometry of $Ti_x(CN)$ compound is close to that of Ti_2C . The formation of Ti_2C -type carbide was noted in [17] which considered to the formation of carbide phases in high alloyed titanium β -alloys. It should be noted that the possibility of the formation of such a carbide in PT-1M alloy at carbon content up to 0.07 wt.% has been confirmed in [18]. According to this study, in Ti-C system at carbon content less than 0.08 wt.% the carbide phase precipitates precisely from α -phase.

The lack of information about the presence of nitrides and carbides in most industrial titanium alloys is probably due to the insignificant concentration of nitrogen and carbon in these alloys. Therefore, it is hard to reveal the nitrides and carbides in the alloy structure by means of microscopy. The other possible reason is that both nitrogen and carbon are dissolved in α - and β -solid solutions within the solubility limit. The latter can be proved by the fact that calculation using ThermoCalc software does not confirm the formation of nitrides in several of investigated alloys (Table 2).

In order to analyze the effect of chemical composition of PT-1M alloy on the phase transformation temperatures such as liquidus temperature ($T_{L\leftrightarrow\beta}$), solidus temperature ($T_{L+\beta\leftrightarrow\beta}$), $\beta\leftrightarrow\alpha+\beta$ - polymorphic transformation temperature ($T_{\beta\leftrightarrow\beta+\alpha}$) it was decided to use a so-called aluminum equivalent (Al_{eq}) similarly to strength equivalent and structural aluminum equivalent [1]. The following aluminum equivalents were suggested to use: $Al_{eq}^{T\beta}$ for $T_{\beta\leftrightarrow\beta+\alpha}$ temperature, Al_{eq}^{Tliq} for $T_{L\leftrightarrow\beta}$ temperature, and Al_{eq}^{Tsol} for $T_{L+\beta\leftrightarrow\beta}$ temperature. The effect of each alloying element in the proposed aluminum equivalents on the corresponding temperature was estimated through equivalent content of aluminum with a certain coefficient. This was proposed based on the binary phase diagrams of Ti-X system, where X is one of alloying elements (Al, Fe, Zr, Si, O, N, C) of the investigated alloy. The coefficient was calculated using the binary phase diagrams [19] and published data [20, 21] proportionally to effect of X element on the corresponding temperature in Ti-X system in comparison to that on aluminum on the same temperature in Ti-Al system.

Data used for calculating $Al_{eq}^{T\beta}$, Al_{eq}^{Tliq} , Al_{eq}^{Tsol} aluminum equivalents are given in Table 3, Table 4.

Table 3. Data for calculating the coefficients of alloying elements for $Al_{eq}^{T\beta}$.

| Element | Temperature of transformation, °C | C_β at transformation, wt. % | Change in T_β temperature at adding of 1 wt. % of element | Calculation coefficient for $Al_{eq}^{T\beta}$ (rounded) | Source |
|---------|-----------------------------------|------------------------------------|---|--|--------|
| Al | — | — | 14.5 | 1 | [20] |
| O | — | — | 200 | 13.8 | [20] |
| N | — | — | 550 | 37.9 | [20] |
| C | — | — | 200 | 13.8 | [20] |
| Zr | 605 | 67.37 | −4.12 | −0.3 | [21] |
| Fe | 595 | 17.07 | −16.84 | −1.2 | [21] |
| Si | 865 | 0.65 | −26.92 | −1.9 | [21] |

The calculation coefficient of alloying elements may assume both positive and negative values. The coefficient assumes positive value if element increases $T_{\beta\leftrightarrow\beta+\alpha}$, $T_{L\leftrightarrow\beta}$, and $T_{L+\beta\leftrightarrow\beta}$ temperatures; the coefficient assumes negative value if element decreases these temperatures. Data reported in [20, 21] were used to calculate $Al_{eq}^{T\beta}$. According to [20], for alloy containing up to 2% Al (wt. %) adding of 1 wt. % of aluminum increases T_β by 14.5°C. This value was taken for normalization of other values. The coefficients for O, N, C were taken from [21]. The coefficients for Zr, Fe, Si were calculated based on data given in [22], minimum temperature of $\beta\leftrightarrow\beta+\alpha$ – polymorphic transformation and corresponding content of element.

Table 4. Data for calculating the coefficients of alloying elements for Al_{eq}^{Tliq} , Al_{eq}^{Tsol} .

| Element | Temperature of liquidus/solidus transformation, °C | Concentration at liquidus/solidus transformation, wt. % | Change of T^{liq}/T^{sol} at adding of 1 wt. % of element | Calculation coefficient for $Al_{eq}^{Tliq}/Al_{eq}^{Tsol}$ | Source |
|---------|--|---|---|---|--------|
| Al | 1700/1697 | 3.0/3.0 | 10.0/9.0 | 1.0/1.0 | [19] |
| O | 1720/1720 | 1.7/2.8 | 28.9/17.7 | 2.9/2.0 | [19] |
| N | 2020/2020 | 1.2/1.9 | 291.7/184.2 | 29.2/20.4 | [19] |
| C | 1645/1645 | 0.5/0.2 | -54.3/-163.4 | -5.5/-18.0 | [19] |
| Zr | 1540/1540 | 50.0/50.0 | -2.6/2.6 | -0.3/-0.3 | [19] |
| Fe | 1085/1085 | 32.8/24.8 | -17.8/-23.6 | -1.8/-2.6 | [19] |
| Si | 1330/1330 | 8.6/2.9 | -39.8/-113.5 | -4.0/-12.6 | [19] |

Based on the coefficients given in Table 3, Table 4 the following formulas for the calculation of $Al_{eq}^{T\beta}$, Al_{eq}^{Tliq} , and Al_{eq}^{Tsol} aluminum equivalents for PT-1M alloy were suggested:

$$Al_{eq}^{T\beta} = C_{Al} + 37.9C_N + 13.8(C_O + C_C) - 1.9C_{Si} - 1.2C_{Fe} - 0.3C_{Zr}, \quad (1)$$

$$Al_{eq}^{Tliq} = C_{Al} + 29.2C_N + 2.9C_O - 5.5C_C - 4.0C_{Si} - 1.8C_{Fe} - 0.3C_{Zr}, \quad (2)$$

$$Al_{eq}^{Tsol} = C_{Al} + 20.4C_N + 2.0C_O - 18.2C_C - 12.6C_{Si} - 2.6C_{Fe} - 0.3C_{Zr}, \quad (3)$$

where C_{Al} , C_N , C_O , C_C , C_{Si} , C_{Fe} , C_{Zr} is the content of Al, N, O, C, Si, Fe, and Zr, correspondingly (wt. %).

The calculated values of $Al_{eq}^{T\beta}$, Al_{eq}^{Tliq} , and Al_{eq}^{Tsol} aluminum equivalents for all investigated alloys are listed in Table 5.

Table 5. Calculated values of aluminum equivalents for different chemical compositions of PT-1M alloy.

| Alloy No | 1 | 2 | 3 | 4 | 5 | 6 |
|--------------------|-------|------|-------|-------|-------|------|
| $Al_{eq}^{T\beta}$ | 0.23 | 5.3 | 4.9 | 0.7 | 2.8 | 2.5 |
| Al_{eq}^{Tsol} | -1.66 | 0.30 | -1.36 | -0.01 | -0.68 | 0.12 |
| Al_{eq}^{Tliq} | -0.52 | 1.74 | 0.99 | 0.23 | 0.61 | 0.69 |

In Figure 2 are shown the relationships between T_β , T_{liq} , T_{sol} temperatures and corresponding $Al_{eq}^{T\beta}$, Al_{eq}^{Tliq} , Al_{eq}^{Tsol} aluminum equivalents established using ThermoCalc data as well as formulas (1), (2), (3) suggested before. The dependence of crystallization interval $T_{liq} - T_{sol}$ (ΔT) on difference of corresponding aluminum equivalents $Al_{eq}^{Tliq} - Al_{eq}^{Tsol}$ ($\Delta Al_{eq}^{Tliq-Tsol}$) was also estimated.

As in can be seen in Figure 2a and Figure 2b, a good linear correlation is observed between T_β , T_{sol} temperatures and corresponding $Al_{eq}^{T\beta}$, Al_{eq}^{Tsol} aluminum equivalents. The dependence obtained for T_β , T_{sol} temperatures of PT-1M alloy was approximated on pure titanium taking $Al_{eq}^{T\beta} = 0$ and $Al_{eq}^{Tsol} = 0$. The difference between the approximated and the experimentally determined values ($T_\beta = 882.5^\circ\text{C}$, $T_{sol} = 1668^\circ\text{C}$) [22] did not exceed 1°C . Calculated T_β value for Alloy 6 (melt composition of PT-1M alloy) was 913°C , which is consistent with T_β value equal 920°C determined by method of testing quenching reported in [5]. In general, the calculated T_β value range $886...949^\circ\text{C}$ for the investigated chemical compositions of PT-1M alloy is consistent with published data of T_β values 890°C [23] and 925°C [9]. The difference between calculated maximum and minimum T_β values for PT-1M alloy is 63°C and corresponds to that of α -titanium alloys. The statistical analysis of T_β values of industrial titanium alloys ingots showed that the difference between calculated maximum and minimum T_β values for VT20 alloy is 65°C .

The correlation between T_{liq} temperature and Al_{eq}^{Tliq} aluminum equivalent (Figure 2c) is not as linear as that observed between T_{β} , T_{sol} and corresponding aluminum equivalents (Figure 2a, Figure 2b). The dependence obtained for T_{liq} and Al_{eq}^{Tliq} of PT-1M alloy was approximated on pure titanium taking $Al_{eq}^{Tliq} = 0$. The approximated value of T_{liq} was 1687°C which exceeds the values by 19°C (Figure 2c).

The dependence character of crystallization interval (ΔT) on difference of corresponding aluminum equivalents ($\Delta Al_{eq}^{Tliq-Tsol}$) is close to linear (Figure 2d). The dependence obtained for ΔT and $\Delta Al_{eq}^{Tliq-Tsol}$ of PT-1M alloy was approximated on pure titanium taking $T_{liq} - T_{sol} = 0$ and $Al_{eq}^{Tliq} - Al_{eq}^{Tsol} = 0$. The approximated value of ΔT exceeds the values by 5°C (see formula on Figure 2d). Thus, it can be concluded that the obtained dependence is quite reasonable.

According to the calculations, the crystallization interval of PT-1M alloy varies from 15°C to 95°C depending on the chemical composition of the alloy. The average value of crystallization interval is 52°C. The calculation results of T_{liq} , T_{sol} , and ΔT values are in a good agreement with that of experimental data reported in [7]. The crystallization interval of titanium alloys may vary from 15°C (for VT1L alloy) to 80°C (for VT21L alloy), and the average value of ΔT of most casting titanium alloys (e.g. VT6L, VT3-1L, VT9L, VT14L) is 60°C, which is consistent the results obtained in the present study.

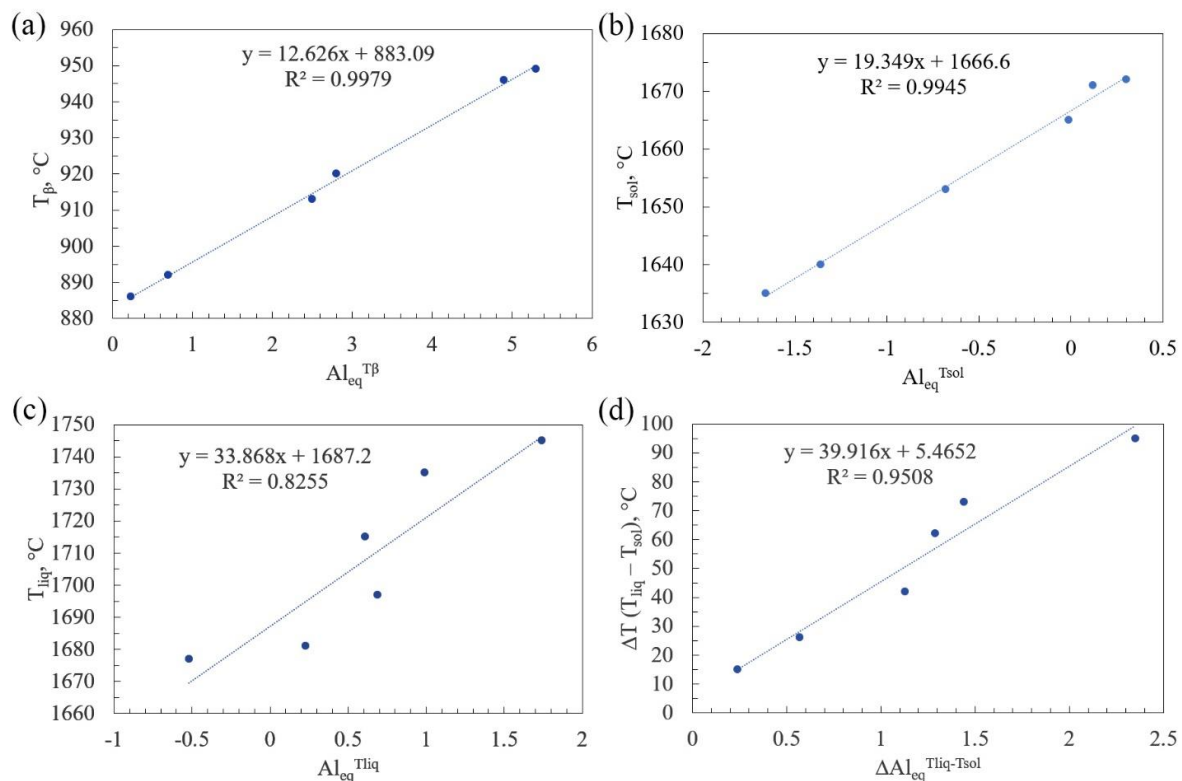


Figure 2. Dependence of T_{β} , T_{sol} , T_{liq} and crystallization interval (ΔT) on $Al_{eq}^{T\beta}$ (a), Al_{eq}^{Tliq} (b), Al_{eq}^{Tsol} (c), $\Delta Al_{eq}^{Tliq-Tsol}$ (d) for PT-1M alloy.

Thus, the dependence of ΔT on $\Delta Al_{eq}^{Tliq-Tsol}$ reported in the present paper can be used to develop and optimize the chemical composition of titanium alloys in order to minimize the crystallization interval ensuring [24] higher casting properties.

Conclusions

1. It has been established the effect of chemical composition and temperature on the phase composition of PT-1M titanium alloy. The possibility of appearance of Ti_2N nitride, $Ti_x(CN)$ -type carbonitride, and Ti_2Fe -type intermetallic in the alloy structure in equilibrium state has been proved by thermodynamic calculations using ThermoCalc software.

2. It has been proposed using of so-called aluminum equivalents (Al_{eq}) for PT-1M alloy to analyze the influence of chemical composition on the phase transformations temperatures change. It has been suggested $Al_{eq}^{T\beta}$ aluminum equivalent for $\beta \leftrightarrow (\beta + \alpha)$ -transformation, $Al_{eq}^{T_{liq}}$ aluminum equivalent for $L \leftrightarrow \beta$ -transformation, and $Al_{eq}^{T_{sol}}$ aluminum equivalent for $(L + \beta) \leftrightarrow \beta$ – transformation as well as calculation formulas:

$$Al_{eq}^{T\beta} = C_{Al} + 37.9C_N + 13.8(C_O + C_C) - 1.9C_{Si} - 1.2C_{Fe} - 0.3C_{Zr}, \quad (4)$$

$$Al_{eq}^{T_{liq}} = C_{Al} + 29.2C_N + 2.9C_O - 5.5C_C - 4.0C_{Si} - 1.8C_{Fe} - 0.3C_{Zr}, \quad (5)$$

$$Al_{eq}^{T_{sol}} = C_{Al} + 20.4C_N + 2.0C_O - 18.2C_C - 12.6C_{Si} - 2.6C_{Fe} - 0.3C_{Zr}, \quad (6)$$

where C_{Al} , C_N , C_O , C_C , C_{Si} , C_{Fe} , C_{Zr} is the content of Al, N, O, C, Si, Fe, and Zr, correspondingly (wt.%).

3. It has been found out the linear correlation dependence between the characteristic temperatures (T_β , T_{liq} , T_{sol} , and ΔT ($T_{liq} - T_{sol}$)) of PT-1M alloy and the proposed aluminum equivalents ($Al_{eq}^{T\beta}$, $Al_{eq}^{T_{liq}}$, and $Al_{eq}^{T_{sol}}$, and $\Delta Al_{eq}^{T_{liq}-T_{sol}}$ ($Al_{eq}^{T_{liq}} - Al_{eq}^{T_{sol}}$).

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